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Tracking the Variable North Atlantic Sink for Atmospheric CO₂

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The oceans are a major sink for atmospheric carbon dioxide (CO₂). Historically, observations have been too sparse to allow accurate tracking of changes in rates of CO₂ uptake over ocean basins, so little is known about how these vary. Here, we show observations indicating substantial variability in the CO₂ uptake by the North Atlantic on time scales of a few years. Further, we use measurements from a coordinated network of instrumented commercial ships to define the annual flux into the North Atlantic, for the year 2005, to a precision of about 10%. This approach offers the prospect of accurately monitoring the changing ocean CO₂ sink for those ocean basins that are well covered by shipping routes.

The natural sinks for atmospheric carbon dioxide have been of great importance in slowing the rate of anthropogenic climate change. Currently, humans emit ~8.5 Pg C year⁻¹ from fossil fuel and cement production, with another ~1.5 Pg C year⁻¹ produced by land use change (1). However, the net increase of the atmospheric concentration is only about half what it would be if all this CO₂ remained in the atmosphere. The remainder is taken up by land vegetation and the ocean in roughly equal measure, as evidenced by simultaneous observations of atmospheric oxygen and CO₂ (2). The ocean uptake of anthropogenically produced CO₂ is reducing the pH of surface waters, an acidification that is expected to have appreciable effects on the marine biota over this century (3).

Both ocean and land uptake is variable in space and time. Estimates of the magnitude and causes of temporal variations averaged over large areas have come from atmospheric observations in inverse models (4–6) and from ocean carbon models (7, 8). The atmospheric inver-

sions suggest that ocean regions such as the North Atlantic and Pacific, as well as continental land regions, exhibit variations in their annual fluxes that are a substantial fraction of their means. Ocean carbon cycle models usually suggest much smaller yearly and decadal changes (7), so it is not clear how much the ocean sinks actually vary. Neither is it clear whether the overall ocean sink is increasing or decreasing: Most models suggest that it should increase with time as atmospheric CO₂ continues to grow, but recent studies have suggested a “saturation” of the sink in the Southern Ocean (5). For the North Atlantic, observations suggest a decrease dating from ~1990 (9), especially between 1995–1996 and 2002–2005 (10). It has been suggested that such variation is linked to the dominant climate mode over the region, the North Atlantic Oscillation (NAO) (8, 10), but the heterogeneous

pattern of CO₂ in the surface ocean has made it difficult to unambiguously identify the nature of this connection. Ocean models forced with atmospheric variation are able to reproduce some of the observational trends, although usually at lower amplitude, and suggest complex patterns of variability within the basins (8).

Although the actual air-sea flux of CO₂ is difficult to measure directly, observations of sea-surface fugacity of CO₂ (fCO₂) can be used to infer it. In recent years, volunteer observing ships (VOS) plying regular routes have been instrumented to make such measurements, and there has been a rapid increase in the quantity of data available (10–14). Figure 1 presents annual flux estimates for the longest-running VOS, for the period 2002 to 2007, between northwestern Europe and the Caribbean [for details of flux calculations, see (10), modified as we describe in (15)]. A substantial variation in the annual fluxes is seen, more than a factor of two over this time period. The observations show decadal rather than interannual variability, with the flux rising and falling over several years. The variation is presumably climatically forced (8, 16), although a relationship with the NAO is not immediately obvious.

The North Atlantic and Pacific are well covered by shipping, and networks of VOS in these oceans might form the backbone of an observing system to continuously monitor the air-sea exchange of carbon dioxide. In 2005, a trial of such a network in the North Atlantic was initiated under CarboOcean, a European Union-funded project, and here we use a year of observations to map the air-sea flux of the region and to evaluate the performance of the network. Figure 2A shows the location of the observations in 2005 on which our evaluation is based. These include VOS routes established specifically for the project (e.g., 1 and 4) and other routes and time series stations of longer establishment.

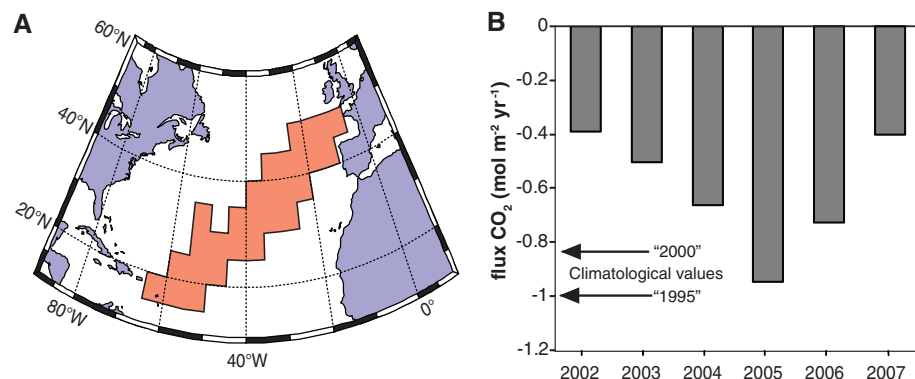


Fig. 1. Annual sea-air fluxes of CO₂ calculated from data on a shipping route between the United Kingdom and the Caribbean. Details of the data collection and the methods of calculating averages and fluxes are given in (10), modified as we describe in (15). (A) Mosaic of 5° by 5° tiles in which data coverage of the UK-Caribbean route is sufficient to calculate annual fluxes over the years 2002 to 2007. (B) Annual average fluxes for the enclosed area. The fluxes are negative (e.g., from air to sea), not only for the region as a whole but for every individual tile within it. Fluxes calculated using “climatological” values of air-sea fCO₂ gradient in this region, referenced to 2000 (14) or 1995 (23), are also indicated. Although these may be indicative of fluxes at these earlier times, they are not strictly applicable to any given year.

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In total, about 125,000 measurements taken in 2005 have gone into this study. Figure 2B shows the locations of observations divided into 2-month periods, to indicate temporal coverage through 2005 (15).

To estimate basinwide fluxes, we constructed maps of sea-surface $f\text{CO}_2$ across the region and through time, by the method of developing relationships between the observed $f\text{CO}_2$ and independent variables, for which data products

were available covering the entire domain. The variables chosen were sea surface temperature (SST) and mixed layer depth (MLD) (15). To obtain a data set of matched parameters from which to begin mappings, these fields were averaged or interpolated onto a 1° by 1° by once-per-day grid, and matched with $f\text{CO}_2$ measurements binned to the same grid, wherever they existed. Observations taken over shelf or shelf-break waters (water depth < 1000 m) were excluded. The rationale for the choice of SST and MLD is that $f\text{CO}_2$ is strongly influenced by temperature change and mixing with subsurface waters, which should be well captured by these variables. Satellite-derived chlorophyll was also initially included (as a proxy for biological activity), but it was found to be of limited utility in predicting $f\text{CO}_2$ and was finally dropped from the analysis. Two different mapping techniques were tested, one based on conventional multivariate linear regressions (MLR) applied after dividing the domain into subregions, and the other based on a self-organizing map (17) covering the entire spatial domain and year with a single map (15).

Fluxes were calculated from the mappings using the gas exchange equation $F = K\alpha\Delta f\text{CO}_2$. Here, $\Delta f\text{CO}_2 = f\text{CO}_{2\text{surface}} - f\text{CO}_{2\text{atm}}$ is the difference between sea-surface and atmospheric $f\text{CO}_2$, K is the gas transfer velocity (parameterized as a function of wind speed and temperature), and α is the solubility of CO_2 in the surface seawater. This approach has historically suffered from a lack of agreement between parameterizations of K derived from in situ measurements and those calibrated for agreement with the global bomb-derived ^{14}C budget. However, recent reanalysis of the ^{14}C budget has largely resolved these discrepancies (18, 19), leading to increased confidence in estimates of CO_2 fluxes by this method. Parameterization of K used National Centers for Environmental Prediction/National

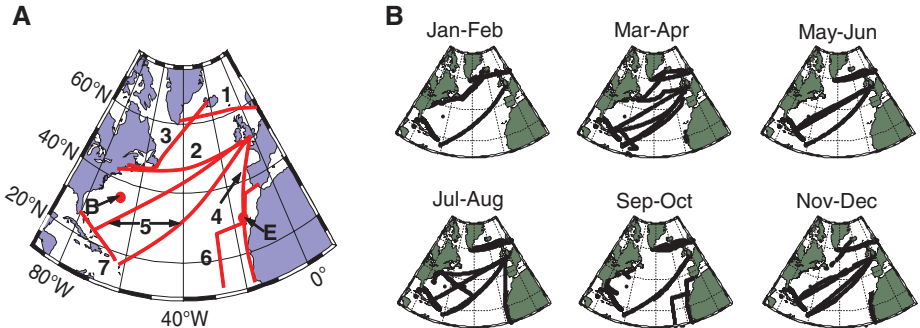


Fig. 2. (A) Location of regular VOS routes supplying data to the Carbo-Ocean network in 2005. Also shown are the locations of time series stations (B, Bermuda Atlantic time series station; E, European station for time series in the ocean, Canary Islands). (B) Post-plot of data binned into 2-month intervals through the year 2005.

Table 1. Integrated net ocean-to-atmosphere flux (10° to 65°N) across the North Atlantic in 2005. Values in the table are normalized to an area of 3.1 by 10^{13} m^2 for comparison (15). P_{CO_2} , partial pressure of CO_2 .

Method of interpolation	Value (Pg C year^{-1})	Area average ($\text{mol m}^{-2} \text{ year}^{-1}$)
MLR in 10° latitude bands	−0.274	−0.737
MLR in 20° latitude bands	−0.251	−0.675
MLR in 30° latitude bands	−0.246	−0.661
Self-organizing mapping	−0.238	−0.640
Mean	−0.252	−0.677
Standard deviation	0.015 (6.1%)	0.040
Using 1995 climatological ΔP_{CO_2} values*	−0.340	−0.914
Using 2000 climatological ΔP_{CO_2} values*	−0.300	−0.806

*Estimates made using climatological maps of ocean-atmosphere CO_2 gradient, which are based on analysis of observations spanning many decades, adjusted to 1995 or 2000 using assumptions described by Takahashi *et al.* (14, 23). These flux estimates used identical gas transfer velocities (e.g., calculated using 2005 winds and temperatures) to the upper rows, so that the differences are due to the different $\Delta f\text{CO}_2$ fields only. For methodological reasons (14), the 1995 climatological value may have overestimated the magnitude of the flux into regions north of 45°N . Both climatological estimates are based on compilations of data collected over decades and are therefore not precise estimates for a given year.

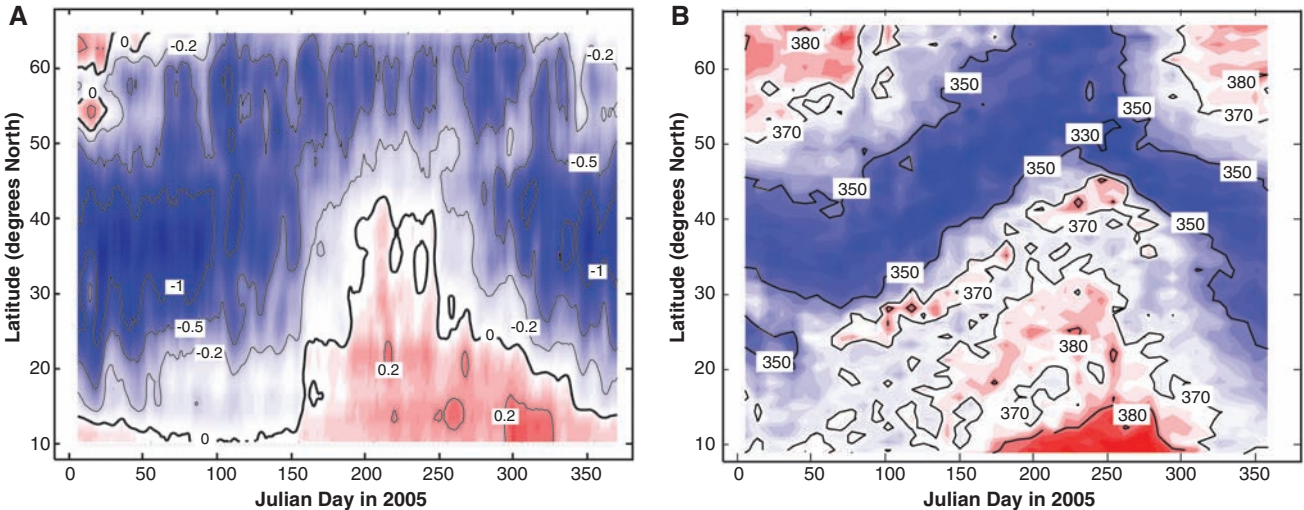


Fig. 3. Contours of ocean-atmosphere flux of CO_2 [(A), in Tmol year^{-1} per degree of latitude] and surface ocean $f\text{CO}_2$ [(B), in μatm] in the North Atlantic in 2005, as a function of latitude and time. Fluxes are positive from sea to air.

The color scheme shades regions of strong positive fluxes and high $f\text{CO}_2$ red, strong negative fluxes and low $f\text{CO}_2$ blue, and near-neutral regions white. The zero flux contour is shown thicker than the others.

Center for Atmospheric Research (NCEP/NCAR) 6-hour winds and surface temperatures (20) and the wind-speed relationship of Nightingale *et al.* (21). The use of winds with 6-hour time resolution captures the great majority of the short-term variation in gas flux (15, 22).

Table 1 shows values of the flux integrated across the North Atlantic throughout the year 2005, for several different methods of mapping. The stability of the calculation gives us some confidence that the overall flux is well constrained by the observations, so is relatively insensitive to the details of the mapping technique. Shown for comparison are flux estimates using climatological values of $f\text{CO}_2$ referenced to 1995 (23) and to 2000 (14), but for the same area and using the identical 2005 gas transfer velocities as the observations, so that differences compared to the observations are entirely due to the $\Delta f\text{CO}_2$ distribution. For the region as a whole, these both show substantially higher sink strengths. Within the restricted area of the time series in Fig. 1, the climatological values are similar to those of 2005 but considerably larger than other years. Taken together, therefore, these comparisons suggest a decline in sink strength from the 1990s to the present decade, but one which is nonuniform with location.

We used two methods to more formally estimate uncertainties: (i) application of geostatistical techniques, proceeding from semivariograms of the residual $f\text{CO}_2$ fields (24) and (ii) application of our methods to $f\text{CO}_2$ fields generated by an eddy-resolving biogeochemical ocean model of the North Atlantic (25). These two techniques have very different underlying assumptions. The second method, in particular, explicitly tests for bias introduced by the mapping techniques. Although the uncertainty on the flux at any given point is in general large (26), it decreases with integration over larger domains. Both our methods of estimating uncertainty indicate that, integrated over the North Atlantic from 10°N to 65°N , the $1-\sigma$ error on the annual mean $f\text{CO}_2$ is $\sim 10\%$ of $\Delta f\text{CO}_2$, with somewhat smaller errors propagating through to the derived flux (15). The broad agreement between two uncertainty analyses having very different underlying assumptions gives us additional confidence in these estimates.

Year-to-year differences are dominated by variability in $\Delta f\text{CO}_2$ (10). Hence, annual flux estimates from different years may also be compared with a precision of $\sim 10\%$. The absolute values, however, have larger uncertainty because of the systematic error arising from the parameterization of gas transfer. Recent estimates for the global gas transfer rate appropriate to CO_2 span a range from 14.6 to $17.1 \text{ cm hour}^{-1}$ (18, 19, 27) and suggest that there remains a $\sim 20\%$ uncertainty here. We therefore quote a flux of $0.25 \pm 0.05 \text{ Pg C}$ for the 10°N to 65°N region for 2005, where the uncertainty is approximately 1σ . Our errors compare favorably with previous attempts to observationally constrain surface-atmosphere fluxes over continents or oceans. For example, the recent North American net “natural” sink,

calculated from land use analyses, is believed to be $0.49 \text{ Pg C year}^{-1}$, with 95% confidence limits ($\sim 2 \sigma$) of 50%, where, however, the estimate is the average over 5- to 10-year periods (28), so that interannual variability cannot be addressed. Estimates made by a dense network of atmospheric observations to provide a “top-down” constraint place this sink in the range of 0.4 to $1.0 \text{ Pg C year}^{-1}$ (29). The VOS network is thus capable of defining the North Atlantic sink to substantially better precision, and somewhat better absolute accuracy, than is currently possible on the land surface.

Figure 3 shows both $f\text{CO}_2$ and air-sea flux integrated across the North Atlantic, as a function of latitude and time through the year (15). The figures show a major sink area in winter due to low $f\text{CO}_2$, extending from 20° to 50°N , which migrates northward through the subpolar gyre during the year. In 2005, it reached a maximum flux into the ocean in March and April, and then weakened through the summer. This CO_2 sink is maintained year-round by the cooling, northward transport of surface waters and is accentuated by the unfolding spring and summer phytoplankton bloom. Also prominent is the subtropical gyre annual cycle driven by temperature increase and strongest in the west. In 2005, the subtropical gyre was a net sink during the winter months but developed into a source as the summer progressed.

These broad features are repeated each year and are seen in climatological estimates of the flux (14, 23), but, it now seems clear, they have substantial interannual and decadal variability around the climatology. Recent work using data before 2005 from some of the Atlantic VOS lines indicates that in 2002 to 2004, the net flux into some areas was only $\sim 50\%$ of mid-1990s values (10, 13). Repeating the flux calculation, but replacing the 2005 $\Delta f\text{CO}_2$ observations with climatological air-sea CO_2 gradients referenced to 2000 (14) or 1995 (23), yields annual sinks of respectively 0.30 or 0.34 Pg C (Table 1). Although the climatological values cannot be unambiguously related to the actual situation in any given year, this suggests that $\Delta f\text{CO}_2$ was on the order of 20% or more lower in 2005 than 5 to 10 years earlier.

Our work demonstrates that an observing network based on commercial VOS is able to constrain the atmosphere-ocean flux of CO_2 into the North Atlantic with good precision. VOS networks are very cost-efficient because the ships are already in place. VOS also operates in the North Pacific and South Atlantic, and a similar approach should in principle be applicable in these oceans, too. An increasing number of moorings in the equatorial Pacific and Atlantic are also now equipped with instrumentation to observe surface $f\text{CO}_2$, so that a greatly improved precision of in situ observation of at least the Northern Hemisphere oceans is now possible. Such an observing system will greatly aid in understanding the ocean sink for atmospheric carbon dioxide and the progress of ocean acidification. It will also provide

a valuable “top-down” constraint on the land sinks, which are more heterogeneous and thus more difficult to observe directly.

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Supporting Online Material

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Materials and Methods
Figs. S1 to S3
Table S1
References

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